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Crosslinkable mixtures, a method of preparing them and their use

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(56) Related Art  
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Crosslinkable mixtures, a method of preparing them and their use

ABSTRACT OF THE DISCLOSURE

A crosslinkable mixture, containing

- a) at least one polysiloxane which contains at least two olefinically or acetylenically unsaturated multiple bonds,
- b) at least one polyhydrogensiloxane which contains at least two hydrogen atoms directly bonded to silicon,
- c) at least one substance which catalyzes hydrosilylation,
- d) at least one inhibitor, and
- e) at least one phosphorus compound of the formula



wherein

R each independently is H, C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl, C<sub>7</sub>-C<sub>31</sub>-alkylaryl, or OR<sup>1</sup>,

R<sup>1</sup> each independently is C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl, C<sub>7</sub>-C<sub>31</sub>-alkylaryl, and

wherein the aryl radicals may optionally be substituted with linear or branched alkyl radicals,

- f) optionally other auxiliary materials

for the coating of a substrate.

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ORIGINAL  
COMPLETE SPECIFICATION  
STANDARD PATENT



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Invention Title:

Crosslinkable mixtures, a method of preparing them and their use

The following statement is a full description of this invention, including the best method of performing it known to me:-

Crosslinkable mixtures, a method of preparing them and their use

5 The present invention relates to crosslinkable mixtures, to a method of preparing them and to their use.

10 The use of silicones for the adhesive coating of substrates such as paper, plastics or metal foils, for example, i.e. for coatings which repel adhesive substances, has been known for some time. In this connection, a liquid mixture comprising a vinyl-containing polydiorganosiloxane, an Si-H-containing polydiorganosiloxane (polyhydrogensiloxane) and a suitable catalyst is applied to the paper and is cured at elevated temperatures by what is termed a hydrosilylation reaction (addition-crosslinking systems). A solid silicone layer which adheres to the substrate is formed in the course of this procedure. The silicone layer is applied to the substrate via rollers, which take up the liquid, reactive silicone mixture from a vat and transfer it to the substrate. If a substrate coating machine such as this breaks down for an extended period of time for technical reasons, the silicone film is capable of crosslinking on the rollers even at room temperature. This results in silicone residues on the rollers which are extremely difficult to remove, and which make it impossible to achieve trouble-free operation again after the machine has been re-started.

20 Since problems which are similar in principle also arise in other applications of addition-crosslinking silicone systems (e.g. LSR), numerous additives (inhibitors) have been developed in the past, the function of all of which is to keep the reactive mixtures liquid for longer at room temperature. Thus US-A-4 329 275, for example, discloses the use of phosphorus compounds in combination with peroxides as working life extenders. However, the addition of peroxides is associated with safety risks during the production and processing of mixtures of this type. US-A-4 645 815 relates to the use of cyclometallized Pt phosphite complexes. In this disclosure the catalyst complex is produced in a separate step, which is associated with additional costs. However, both of the aforementioned possible solutions have a further, very serious disadvantage. They were developed for systems which employ relatively low concentrations of catalyst (10 ppm). Silicone mixtures which have a considerably higher content of catalyst (100 ppm) are used for the coating of substrates, however. In systems with high catalyst concentrations such as this, the aforementioned inhibitors do not fulfil the requirements which are desired by the market as regards room temperature

5 stability. US-A-4 851 452 proposes the use of Pt-vinylsiloxane complexes in combination with Pt-phosphine complexes as catalysts which have a significantly decreased activity at room temperature. However, phosphines have the disadvantage that the activity of the catalyst is also decreased to an appreciable extent at elevated temperatures ( $T=180^{\circ}\text{C}$ ), i.e. under curing conditions. For this reason, they are only of very limited value for substrate coating applications.

10  $\alpha$ -hydroxyacetylenes, which are described in detail in US-A-3 445 420, constitute another group of inhibitors which can be used. These compounds do in fact prolong the working life of the system "in bulk", but are extremely unsatisfactory as regards prolonging the working life of thin films such as those which are present on the rollers during the coating operation.

15 The object of this invention is therefore to provide suitable mixtures which decrease the activity of the catalyst at room temperature, even in rapid, addition-crosslinking silicone systems, without prolonging the curing times under reaction conditions. At the same time, an object is to be able to prepare the mixture in a simple manner and without safety risks.

20 It has now been found that these problems can be solved with addition-crosslinking polysiloxane mixtures which contain Pt compounds or elemental Pt, an  $\alpha$ -hydroxyacetylene or an ester of a dicarboxy acid and at least one organophosphorus compound of the type specified in detail below.

The present invention therefore relates to crosslinkable mixtures containing

- 25
- a) at least one polysiloxane which contains at least two olefinically or acetylenically unsaturated multiple bonds,
  - b) at least one polyhydrogensiloxane which contains at least two hydrogen atoms directly bonded to silicon,
  - c) at least one substance which catalyzes hydrosilylation,
  - d) at least one inhibitor, and
  - e) at least one phosphorus compound of formula (I):



wherein

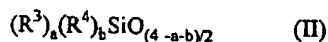
R each independently is H, C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl, C<sub>7</sub>-C<sub>31</sub>-alkylaryl, or OR<sup>1</sup>

5 R<sup>1</sup> each independently is C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl, C<sub>7</sub>-C<sub>31</sub>-alkylaryl, and

wherein the aryl radicals may optionally be substituted with linear or branched alkyl radicals and R and R<sup>1</sup> may have a different meaning within the molecule, and

10 f) optionally other auxiliary materials.

Component a) in the sense of the invention is preferably a cyclic, linear or branched polysiloxane which is synthesized from units of the general formula (II)



15 Here, R<sup>3</sup> represents a C<sub>2</sub>-C<sub>8</sub> alkenyl radical, e.g. vinyl, allyl, 1-butenyl, 1-hexenyl, etc. The alkenyl radicals may be bonded to silicon atoms within the chain or at the end of the chain. R<sup>4</sup> is a monovalent, saturated hydrocarbon radical containing up to 10 carbon atoms, from the group comprising substituted and unsubstituted alkyl, aryl and arylalkyl radicals. Examples of monovalent radicals R<sup>4</sup> include methyl, ethyl, propyl, isopropyl, butyl, octyl, etc., cyclobutyl, cyclopentyl, 20 cyclohexyl, etc., phenyl, tolyl, xylyl, naphthyl, etc., and benzyl, phenylethyl or phenylpropyl. The relationships  $0 \leq a \leq 3$ ,  $0 \leq b \leq 3$  and  $0 \leq a + b \leq 4$  apply to the integers a and b. a is preferably equal to 0 or 1. In radicals R<sup>4</sup> of the present invention, some or all of the hydrogen atoms may be substituted by fluorine and / or chlorine, bromine, or iodine atoms or cyano radicals. This means 25 that R<sup>4</sup> may also be a chloromethyl, trifluoropropyl, chlorophenyl, dibromophenyl, β-cyanoethyl, β-cyanopropyl or γ-cyanopropyl radical, for example.

Using the nomenclature familiar to one skilled in the art:

- 5 M:  $(\text{CH}_3)_3\text{SiO}_{1/2}$   
 D:  $(\text{CH}_3)_2\text{SiO}_{2/2}$   
 T:  $(\text{CH}_3)\text{SiO}_{3/2}$   
 $\text{M}^{\text{Vi}}$ :  $(\text{CH}_2=\text{CH})(\text{CH}_3)_2\text{SiO}_{1/2}$   
 $\text{D}^{\text{Vi}}$ :  $(\text{CH}_2=\text{CH})(\text{CH}_3)\text{SiO}_{2/2}$

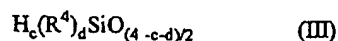
the following examples of component a) can be cited:

- $\text{M}_2\text{D}_{100}\text{D}_3^{\text{Vi}}$   
 $\text{M}_2^{\text{Vi}}\text{D}_{180}$   
 $\text{M}^{\text{Vi}}\text{MD}_{100}\text{D}_3^{\text{Vi}}$   
 10  $\text{T}_5\text{D}_{550}\text{M}_7^{\text{Vi}}$   
 $\text{T}_3\text{D}_{500}\text{M}_2^{\text{Vi}}\text{M}_3$   
 $\text{T}_6\text{D}_{300}\text{D}^{\text{Vi}}\text{M}_4^{\text{Vi}}\text{M}_4$

The molar proportion of unsaturated radicals of type  $\text{R}^3$  may be selected to have any desired value.

- 15 In component a), the molar proportion of unsaturated radicals of type  $\text{R}^3$  should preferably be between 0.01 and 10 mmole per gram, more preferably between 0.05 and 1 mmole per gram, and most preferably between 0.1 and 0.7 mmole per gram of component a). The viscosity of component a) is preferably between 0.01 and 100,000 Pa·s, more preferably between 10 and 100,000 mPa·s at 25°C.

- 20 Component b) in the sense of the present invention is a polysiloxane which is synthesized from units of general formula (III)

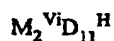
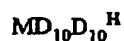


- 25 where  $\text{R}^4$  has already been defined above and may optionally also have the meaning of  $\text{R}^3$ . The stoichiometric suffices c and d are integers, where  $0 \leq d \leq 3$  and  $0 \leq c \leq 2$ , and where  $0 \leq c + d \leq 4$ . Preferably,  $0 \leq c \leq 1$ .

Using the nomenclature familiar to one skilled in the art:



the following examples of component b) can be cited:



10 (where M, D,  $M^{Vi}$  and  $D^{Vi}$  are defined as for component a)).

The molar proportion of hydrogen atoms which are directly bonded to a silicon atom in component b) may be selected to have any desired value.

15 In component b), the molar proportion of hydrogen atoms which are directly bonded to a silicon atom is preferably between 0.01 and 17 mmole, more preferably between 0.1 and 17 mmole, and most preferably between 1 and 17 mmole per gram of component b).

20 In the total mixture described above, components a) and b) should preferably be present in a quantitative ration such that the molar ratio of hydrogen atoms which are directly bonded to silicon atoms (SiH) in component b) to the unsaturated radicals (Si-vinyl) in component a) is between 0.01 and 300, more preferably between 0.05 and 20, even more preferably between 0.5 and 10 and most preferably between 1 and 5.

25 Component c) in the sense of the invention preferably comprises the elements platinum, rhodium, iridium, nickel, ruthenium and/or palladium, in elemental form on a supporting substance or in the form of their compounds. Platinum compounds or platinum complexes are preferred, such as  $H_2PtCl_6$ , platinum-olefin complexes, platinum-alcohol complexes, platinum-vinylsiloxane complexes and





also elemental platinum on a supporting substance, such as activated carbon,  $\text{Al}_2\text{O}_3$  or  $\text{SiO}_2$  for example. Component c) is most preferably a platinum-vinyl-siloxane complex. Platinum-vinylsiloxane complexes most preferably contain at least two olefinically unsaturated double bonds in the siloxane; see US 3 715 334 for example.

The term "siloxane" includes polysiloxanes, i.e. it includes vinylpolysiloxanes also, for example. The proportion of component c) in the total mixture should preferably be between 1 and 1000 ppm, more preferably between 1 and 500 ppm, and most preferably between 25 and 250 ppm.

10 Component d) in the sense of the invention comprises all the inhibitors which are known in the prior art, such as maleic acid and derivatives thereof, amines, alkyl isocyanurates and acetylenically unsaturated alcohols in which the OH group is bonded to a carbon atom adjacent to the C-C triple bond, such as those described in detail in US 3 445 420, for example. Component d) is preferably 2-methyl-3-butyn-2-ol or 1-ethynylcyclohexanol or ( $\pm$ ) 3-phenyl-1-butyn-3-ol. Component d) is preferably added to the mixture in a quantitative proportion of 0.0001 % to 5 % with respect to the total weight of mixture, more preferably 0.01 % to 2 %, and most preferably 0.1 to 1 %.

20 Component e) in the sense of the invention is an organophosphorus compound of type  $\text{PR}_3$ . In this respect, R is a hydrogen atom or a monovalent hydrocarbon radical from the group comprising  $\text{C}_1$ - $\text{C}_{18}$  alkyl,  $\text{C}_6$ - $\text{C}_{30}$  aryl and  $\text{C}_7$ - $\text{C}_{31}$  alkylaryl. R may have a different meaning within a molecule. In a preferred embodiment of the present invention, component e) is a compound of formula  $\text{P}(\text{OR}^1)_3$ , where  $\text{R}^1 = \text{C}_1$ - $\text{C}_{18}$  alkyl,  $\text{C}_6$ - $\text{C}_{30}$  aryl or  $\text{C}_7$ - $\text{C}_{31}$  alkylaryl, wherein the aryl radicals may optionally be substituted with linear or branched alkyl radicals and  $\text{R}^1$  may have different meanings within the molecule. Examples of component e) include triphenylphosphine, triethylphosphine, tributylphosphine, phosphoric acid trimethyl ester, phosphoric acid triisopropyl ester, phosphoric acid triphenyl ester or tris-(2,4-di-tert. butylphenyl)-phosphite. Component e) of the invention can also include phosphite derivatives of pentaerythritol, such as, for example, bis (2,4-di-tert.butylphenyl)-pentaerythritol diphosphite (obtainable from the Greck Dakes Company under the trade name Alkanox P24®). Component e) of the present invention is preferably added in a quantitative proportion of 0.0001 % to 5 % with

respect to the total weight of the mixture, more preferably 0.001 % to 4 % and most preferably between 0.001 % and 2.5 %.

5 Examples of auxiliary materials (component f)) in the sense of the invention include polysiloxane resins which are synthesized from components of general formulae (II) and (III), fillers which have a positive effect on the mechanical and electrical properties of the cured mixture according to the invention, such as pyrogenic and precipitated hydrated silicas having a BET specific surface of 50 to 500 m<sup>2</sup>/g, for example. The surfaces of fillers of this type may be modified, e.g. with organosilicon compounds. Modification may also be effected during their  
10 incorporation into the polymer, by the addition of hexamethyldisilazane or 1,3-divinyl-1,1,3,3-tetramethyldisilazane with the addition of water, for example.

Substances such as diatomaceous earths, finely divided quartz flour, amorphous hydrated silicas or carbon blacks may also be used as fillers, for example.

15 In another embodiment the mixtures according to the invention additionally contain water or an organic solvent.

In a preferred embodiment of the invention, components a) and b) are present in a quantitative ratio such that

the SiH:Si-vinyl ratio is between 0.01 and 300,  
the content of component c) is between 1 and 1000 ppm,  
20 the content of component d) is between 0.0001 and 5 %, and  
the content of component e) is between 0.0001 and 5 %.

wherein the quantitative data are given with respect to the total weight of the mixture in each case.

25 This invention also relates to a method of preparing the crosslinkable mixtures according to the invention. In this process, components a), d) and e) are preferably mixed, and thereafter component b) and finally component c) are added.

This invention further relates to the use of the crosslinkable mixture according to the invention for the coating of substrates.



The following examples serve to explain the invention. The invention is not restricted to these examples, however.

### Examples of implementation

5 In the following examples, all parts by weight and percentages are given with respect to the weight of the total mixture unless defined otherwise.

### General experimental procedure for Examples 1a to 1j

The quantitative data which do not appear in this description, as well as the type of compounds and the experimental results, are given in Table 1.

10 9.15 g of a branched polydimethylsiloxane with a viscosity of  $\eta = 250$  mPa·s and a content of vinyl groups bonded directly to silicon of 0.32 mmole/g were mixed optionally with an  $\alpha$ -hydroxyacetylene (see Table 1) and optionally with an organophosphorus compound (see Table 1). 0.75 g of a polyhydrogensiloxane having a content of hydrogen atoms bonded directly to silicon of 11.3 mmole/g were then added. Finally, a sufficient amount of a solution of a Pt-vinylsiloxane  
15 complex were added to the vinyl-containing polymer described above in order to obtain a Pt content of 100 ppm. The meanings of the abbreviations in the following Table are as follows:

	ECH :	1-ethynylcyclohexanol
	PhB :	( $\pm$ )-3-phenyl-1-butyne-3-ol
20	PoV1:	triphenyl phosphite
	PoV2:	triethyl phosphite
	PoV3:	triisopropyl phosphite
	PoV4:	tris-(2,4-di-tert. butylphenyl)-phosphite
	PoV5:	triphenylphosphine oxide (comparison)
25	PoV6:	trimethyl phosphite

**Table 1**

Ex. No.	$\alpha$ -hydroxy-acetylene	Amount	Organo-phosphorus compound	Amount	Working life as thin film	Working life in bulk
1a <sup>1)</sup>	ECH	27 mg	-	-	20 min	24 h
1b <sup>1)</sup>	-	-	PoV4	10 mg	5 s	5 s
1c	ECH	27 mg	PoV4	10 mg	22 h	>10 days
1d	ECH	27 mg	PoV3	10 mg	3 h 15 min	>6 days
1e	ECH	27 mg	PoV2	10 mg	4 h 15 min	>6 days
1f	ECH	27 mg	PoV6	10 mg	2 h 20 min	>6 days
1g <sup>1)</sup>	-	-	PoV1	10 mg	1 h 40 min	>6 days
1h	ECH	27 mg	PoV1	10 mg	22 h	>10 days
1i <sup>1)</sup>	PhB	10 mg	-	-	20 min	24 h
1j	PhB	10 mg	PoV1	10 mg	26 h	>10 days

<sup>1)</sup> comparative examples

The combination of components d) and e) is an essential part of this invention. As can be seen from comparative examples 1a, 1b, 1g and 1i, the presence of only one of the two components does not result in a long working life.

**Example 2**

The following example demonstrates the effect of certain organophosphorus compounds on the curing times at  $T = 180^{\circ}\text{C}$ . An  $\alpha$ -hydroxyacetylene and an organophosphorus compound, of the type and in the amounts given in detail in Table 2, were added to 8.96 g of the vinyl-containing polymer described in Example 1. Thereafter, 0.21 g of the polyhydrogensiloxane described in Example 1 and a sufficient amount of the catalyst solution described in Example 1 were added, in order to obtain a Pt content of 100 ppm. The results are given in Table 2.

**Table 2**

$\alpha$ -hydroxy-acetylene	Amount	Organo-phosphorus compound	Amount	Working life as thin film	Curing time at T = 180°C
ECH	27 mg			20 min	7 s
ECH	27 mg	PoV4	10 mg	21 h	7 s
ECH	27 mg	PoV5	10 mg	50 min	>60 s

It will be understood that the specification and examples are illustrative but not limitative of the present invention and that other embodiments within the spirit and scope of the invention will suggest themselves to those skilled in the art.

**Example 3**

The following example is intended to demonstrate the synergistic effect of the inhibitory activity of a dicarboxylic acid ester and an organophosphorus compound.

9.15 g of a branched polydimethylsiloxane with a viscosity of  $\eta = 250$  mPa·s and a content of vinyl groups bonded directly to the silicon of  $0.32 \text{ mmol g}^{-1}$  are mixed with an organophosphorus compound and a dicarboxylic acid ester. Then 0.55 g of a polyhydrogensiloxane which has a content of hydrogen atoms directly bonded to the silicon of  $11.3 \text{ mmol g}^{-1}$  is added. Then a sufficient quantity of a solution of a Pt/vinylsiloxane complex is added in order to obtain a Pt content of 100 ppm. The exact quantities and the type of inhibitors used are shown in Table 3. The organophosphorus compound has the name given in Example 1.

**Table 3**

Ex. No.	Dicarboxylic acid ester	Quantity	Organo-phosphorus compound	Quantity	Working life as thin film
3a <sup>1</sup>	dimethyl maleate	0 mg	PoV4	10 mg	5 s <sup>2</sup>
3b <sup>1</sup>	dimethyl maleate	32 mg	PoV4	0 mg	25 min
3c	dimethyl maleate	32 mg	PoV4	10 mg	12 h

<sup>1</sup> comparative example

<sup>2</sup>The whole mixture gelled immediately after the introduction of the Pt catalyst solution

Throughout this specification and the claims which follow, unless the context requires otherwise, the word "comprise", and variations such as "comprises" or "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integers or steps.



The Claims defining the invention are as follows:

1. A crosslinkable mixture, containing
- a) at least one polysiloxane which contains at least two olefinically or acetylenically unsaturated multiple bonds,
  - b) at least one polyhydrogensiloxane which contains at least two hydrogen atoms directly bonded to silicon,
  - c) at least one substance which catalyzes hydrosilylation,
  - d) at least one inhibitor, and
  - e) at least one phosphorus compound of general formula (I):



wherein

R each independently is H, C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl, C<sub>7</sub>-C<sub>31</sub> alkylaryl, or OR<sup>1</sup>,

R<sup>1</sup> each independently is C<sub>1</sub>-C<sub>18</sub>-alkyl, C<sub>6</sub>-C<sub>30</sub>-aryl or C<sub>7</sub>-C<sub>31</sub>-alkylaryl, and

wherein the aryl radicals may optionally be substituted with linear or branched alkyl radicals

- f) optionally other auxiliary materials.

2. A crosslinkable mixture according to claim 1, wherein c) is a Pt compound, platinum complex, or elemental platinum on a supporting substance.

3. A crosslinkable mixture according to claim 1, wherein c) is a platinum-vinylsiloxane complex in which the siloxane contains at least two olefinically unsaturated double bonds.
- 5 4. A crosslinkable mixture according claim 1, wherein d) is 2-methyl-3-butyne-2-ol, 1-ethynylcyclohexanol or (±)-3-phenyl-1-butyne-3-ol.
5. A crosslinkable mixture according to claim 1, wherein e) is a compound of the formula  $P(OR^1)_3$ , wherein
 

10  $R^1$  each independently is  $C_1-C_{18}$ -alkyl,  $C_6-C_{30}$ -aryl or  $C_7-C_{31}$ -alkylaryl, wherein the aryl radicals may optionally be substituted with linear or branched alkyl radicals.
6. A crosslinkable mixture according to claim 1, containing water.
7. A crosslinkable mixture according to claim 1, additionally containing an organic solvent.
- 15 8. A crosslinkable mixture according to claim 1, wherein a) and b) are present in a quantitative ratio such that the SiH:Si-vinyl ratio is between 0.01 and 300, the content of component c) is between 1 and 1000 ppm, the content of component d) is between 0.0001% and 5%, and the content of component e) is between 0.0001% and 5%, wherein the quantitative data in each instance are with respect to the total weight of the mixture.
- 20 9. A crosslinkable mixture according to claim 1, which comprises mixing a), d) and e), and subsequently adding b) and finally adding c).
- 25 10. A crosslinkable mixture according to claim 8, wherein c) is a platinum-vinylsiloxane complex in which the siloxane contains at least two olefinically unsaturated double bonds, d) is 2-methyl-3-butyne-2-ol, 1-ethynylcyclohexanol or (±)-3-phenyl-1-butyne-3-ol, e) is a compound of the formula  $P(OR^1)_3$ , wherein
 

$R^1$  each independently is  $C_1-C_{18}$ -alkyl,  $C_6-C_{30}$ -aryl,  $C_7-C_{31}$ -alkylaryl, and





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additionally containing at least one of water and an organic solvent.

11. A method of coating of a substrate by applying thereto a silicone composition comprising a mixture according to any one of claims 1 to 10.
- 5 12. A cross linkable mixture substantially as hereinbefore described in any one of Examples 1 to 3.

DATED this 6th day of January 2000,

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**GE BAYER SILICONES GMBH & CO. KG**

By their Patent Attorneys  
DAVIES COLLISON CAVE

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